From Molecular Radicals to 2D Quantum materials: 2D Covalent Organic Radical Frameworks (CORFs)

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A few years ago, we proposed using stable triarylmethyl radicals as novel open-shell building blocks for 2D covalent organic *radical* frameworks (2D CORFs) [1,2]. When forming a hexagonal lattice, we calculated that 2D hex-CORFs would exhibit antiferromagnetic (AF) ground states and would have energetically close lying closed-shell quinoidal and graphene-like semimetallic states [2]. Soon after this prediction, the first 2D hex-CORF was synthesised and was indeed found to show an AF state [3]. The AF state in 2D hex-CORFs is associated with a parent correlated Mott insulating phase in which unpaired spin-carrying electrons are localised on the radical nodes of the framework [2]. From this parent AF state, graphene-like semimetallic and closed-shell quinoidal/dimerised states should be accessible by modest out-of-plane compression [4] or in-plane strain [5], respectively. Recently, we have studied partial chemical substitution of the spin-carrying C centres in 2D hex-CORFs by B or N. The resulting materials can be viewed as 2D extensions of neutral mixed valence compounds and exhibit emergent spin-frustrated triangular lattices which have the potential to host a range of exotic quantum states (e.g. superconductivity, spin-liquid, novel magnetic ordering) [6].

Overall, our findings establish 2D hex-CORFs as a new class of 2D multifunctional quantum materials that can be tuned by stress/strain/substitution for potential future technological applications.

References

[1] I. Alcón, D. Reta, I. de P. R. Moreira, S. T. Bromley, *Chem. Sci.* 8 (2017) 8, 1027.
[2] I. Alcón, F. Viñes, I de P. R. Moreira, S. T. Bromley, *Nat. Commun.* 8 (2017) 1957.
[3] S. Wu, M. Li, H. Phan, D. Wang, T. S. Herng, J. Ding, Z. Lu, J. Wu, *Angew. Chem. Int. Ed.* 57 (2018) 8007.
[4] R. Santiago, I. Alcón, J. Ribas-Arino, M. Deumal, I. de P. R. Moreira, S. T. Bromley, *Adv. Funct. Mater.* 31 (2021) 202004584.
[5] I. Alcón, R Santiago, J. Ribas-Arino, M. Deumal, I. de P. R. Moreira, S. T. Bromley, *Nat. Commun.* 12 (2021) 1705.
[6] I. Alcón, J. Ribas-Ariño, I. de P.R. Moreira, and S. T. Bromley, *J. Am. Chem. Soc.* (2023) doi.org/10.1021/jacs.2c11185.

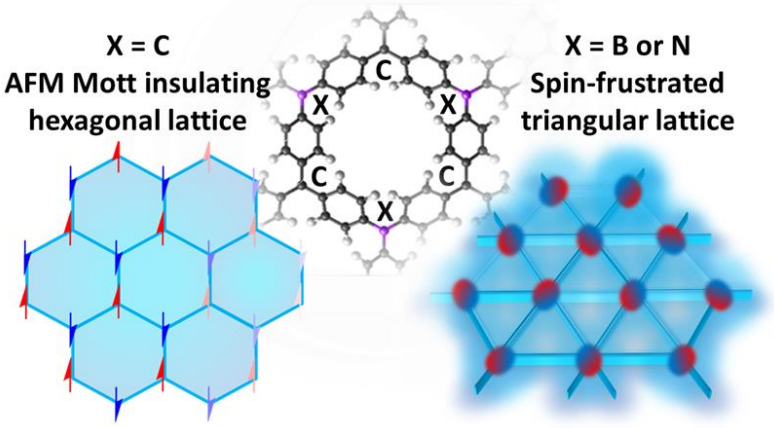

Figure 1: All-carbon 2D hex-CORFs tend to be half-filled correlated Mott insulators (left). Substituting half the carbon centres with B or N leads to emergent 2D spin-frustrated triangular lattices having promise as a platform for exotic quantum materials (right).